• *Invited Letter*

Dynamic Plastic Deformation (DPD): A Novel Technique for Synthesizing Bulk Nanostructured Metals

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While some superior properties of nanostructured materials (with structural scales below 100 nm) have attracted numerous interests of material scientists, technique development for synthesizing nanostructured metals and alloys in 3-dimensional (3D) bulk forms is still challenging despite of extensive investigations over decades. Here we report a novel synthesis technique for bulk nanostructured metals based on plastic deformation at high Zener-Hollomon parameters (high strain rates or low temperatures), *i.e.*, dynamic plastic deformation (DPD). The basic concept behind this approach will be addressed together with a few examples to demonstrate the capability and characteristics of this method. Perspectives and future developments of this technique will be highlighted.

Synthesizing bulk nanostructured metals and alloys (with structural scales below 100 nm) is crucial not only for fundamental researches of the nature of this new class of advanced materials but also for their technological applications in industries as well. Thorough understanding of the intrinsic structure-property relationship (especially for mechanical properties) of nanostructured materials is highly depending upon reliable experimental measurements of bulk specimens. However, a proper synthesis technique for 3-dimensional (3D) bulk nanostructured metals and alloys is still lacking so far, which becomes a bottleneck for development of this material family.

Over the past decades, several techniques have been developed for producing bulk specimens of nanostructured metals, including gas-phase condensation and consolidation[1], consolidation of ball-milled powders[2], electrodeposition[3], crystallization of amorphous solids[4], and various severe plastic deformation (SPD) techniques[5]. Nevertheless, serious drawbacks of these methods exclude their widespreading applications, such as process complications, high processing costs, process-induced contaminations or porosities, or limited kinds of processable materials, *etc.* Among these methods, SPD techniques have received most attentions in recent years owing to their capability of producing 3D bulk specimens and their potentials in technological applications. Yet, SPD techniques are suffered from large grain sizes in the processed samples, usually in the submicron scale (>100 nm). Therefore, bulk samples processed by SPD techniques are frequently referred to as ultrafine-grained (UFG) materials. Of course, properties of UFG materials differ significantly from those of real nano-grained counterparts.

In this letter, we report a novel synthesis technique for producing 3D bulk nanostructured metals by means of plastic deformation at high strain rates, *i.e.*, dynamic plastic deformation (DPD). With this, bulk specimens of several metals and alloys have been successfully produced in which structural scales (grain sizes or twin lamellar thicknesses) are below 100 nm. Superior mechanical properties have been obtained in these bulk nanostructured metals in comparison with those of the UFG samples.

It is well-known that plastic deformation induces grain refinement in metals and alloys. Substantial refinement of grains down to the sub-micrometer regime (>100 nm) in a large number of metallic materials has been achieved *via* plastic deformation with conventional strain rates ($<10^0$ s^{-1}) and at ambient temperature with a large plastic strain (or frequently called severe plastic deformation). With increasing strain, grain sizes decrease and tend to a saturated value of several hundred nanometers for most metals and alloys. For most fcc and bcc metals deformed at low strain rates and at ambient temperature, dislocation activities are dominating deformation mechanism. Hence, strain-induced refinement of grains is realized *via* dislocation activities: dislocation manipulation, rearrangement, formation of various substructures in original coarse grains such as dense dislocation walls (DDWs), dislocation cells, tangles, *etc.* Interaction between these substructures and more dislocations results in formation sub-boundaries of small misorientations, subdividing the original coarse grains into refined blocks. Further straining leads to increasing misorientations, eventually forming randomly oriented refined grains[6].

Obviously, the saturated (or minimum) grain sizes upon increasing straining are controlled by the saturated (maximum) dislocation density, or by the balance between dislocation manipulation and annihilation. When the size of substructures is reduced to a certain value, the generation rate of dislocations induced by straining is equal to the annihilation rate of dislocations at a given strain rate and temperature. Then, dislocation density stored in the material will not increase further with increasing strain, resulting in a saturated size of subgrains or grains. For example, grain sizes of pure Cu can be reduced by extending plastic deformation, reaching a saturated value of about 200 nm with various SPD techniques[7,8]. The corresponding dislocation density in the heavily deformed Cu can be elevated to a value as high as in the order of 10^{15} m^{-2}. This dislocation density can not be increased further so that the saturated grain

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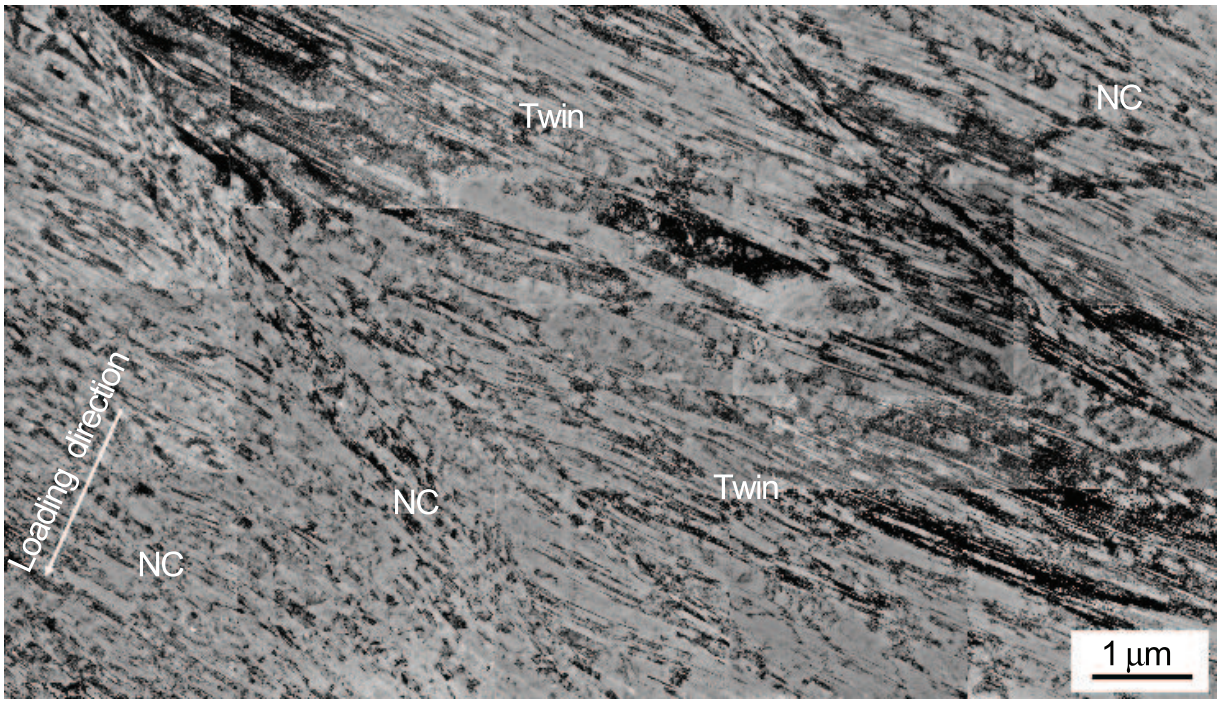

Fig.1 A typical cross-sectional TEM image for the LNT-DPD Cu sample with a plastic strain of ε=2.1 (NC: nanocrystalline)[15]

size can not be reduced by increasing strains at conventional strain rates and ambient temperature. In other metals, a similar phenomenon is observed. The saturated grain sizes for Ni, Al, and Fe are 100 nm, 500 nm, and 100 nm, respectively.

To reduce grain size further below this limit, one has to enhance dislocation storage by either increasing the generation rate of dislocations or suppressing the annihilation (or recovery) kinetics, or both. Dislocation activities are thermally-activated processes of which the kinetics is sensitive to strain rate and temperature. It is known that the dislocation recovery kinetics is effectively suppressed at high strain rates and/or at low temperatures in most metallic materials[9]. Density of the stored dislocations is elevated and the size of substructures is decreased in the deformed samples at high rates and/or low temperatures. Consequently, plastic deformation at high strain rates and/or at low temperatures, referring to as dynamic plastic deformation (DPD), would possibly be an effective approach to refining grains below 100 nm. This concept has been testified by plenty of experimental evidences that nano-sized grains were produced in topmost surface layers in various metals and alloys after surface mechanical attrition treatment (SMAT)[10–13]. Formation of the nano-sized grains originates from the high strain rates (up to 10^{-3} s^{-1})[10–12].

In some metals with low or medium stacking fault energies (SFEs) such as Cu, plastic deformation is accommodated by means of two competitive mechanisms, *i.e.*, dislocation slip and deformation twinning. At high strain rates or at low temperatures when dislocation activities are suppressed, deformation twinning becomes more significant in carrying plastic strain[9]. Twinning occurs near grain boundaries or other defects when the local stress exceeds the critical shear stress for initiating deformation twinning. Formation of multiple twins provides an alternative mechanism for grain refinement in deformed metals. When deformation twin density is high enough, the thicknesses of twins and matrix can be reduced into nanometer scale, *i.e.*, forming nano-scale twin/matrix lamellae. In subsequent deformation, interactions between dislocations and twin boundaries lead to evolution of nano-scale twin/matrix lamellae into nano-sized grains. This mechanism was also observed in the SMAT Cu samples. Grains of about 15 nm in size were formed in the topmost surface layer, originating primarily from fragmentation of nano-scale twins[12]. The nano-scale twins were found to significantly facilitate grain refinement kinetics in the nanometer scale.

In our preliminary experiments that bulk pure Cu specimens were deformed (compressed) at a high strain rate of about 10^3 s^{-1} at liquid nitrogen temperature (LNT)[14–16], *i.e.*, DPD at cryogenic temperatures, multiple deformation twinning becomes a predominant deformation mechanism of the bulk sample. The twin/matrix lamellae thickness is as small as about 45 nm. Formation of nano-scale deformation twins in the bulk DPD Cu samples is distinct from that in the SPD Cu samples deformed at low strain rates and ambient temperatures, in which few deformation twins are formed. In subsequent deformation, a fraction of nano-scale twin structures are evolved into nano-sized grains by means of fragmentation of nano-scale twin/matrix lamellae and shear banding of the twin bundles. Figure 1 shows a typical TEM observation of the as-DPD Cu sample at LNT, which is characterized by nano-scale twin bundles with an average twin/matrix lamellar spacing of about 45 nm (about 35% in volume) embedded in nano-grained matrix with an average size of about 66 nm. Detailed analysis of the nano-grain formation

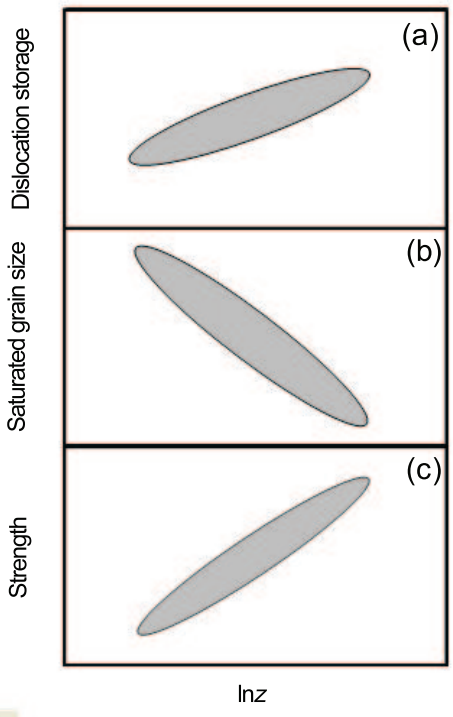

Fig.2 Schematic of variations of stored dislocation density (a), the saturated grain size (b), and strength (c) with Zener-Hollomon parameter ($\ln Z$) in deformed materials

in the DPD Cu sample can be found in literature [15]. With the similar DPD process, nanostructures (nano-sized grains and nano-scale twins) in several Cu-Al and Cu-Zn alloys (brass) have been successfully synthesized in the authors′ laboratory[17,18].

Increasing strain rates is equivalent to decreasing deformation temperature in suppression of dislocation activities. The Zener-Hollomon parameter (Z) incorporates strain rate ($\dot{\varepsilon}$) and temperature (T) into a unified expression which contains the activation energy for diffusion (Q)[19]:

$$\ln Z = \ln\dot{\varepsilon} + Q/RT \tag{1}$$

Clearly, Z parameter increases with either increasing strain rate or decreasing temperature. For example, the Zener-Hollomon parameter corresponding to deforming Cu at a strain rate of 10^{-3} s^{-1} at ambient temperature, is $\ln Z$=21–23. But in a DPD process at a rate of 10^{3} s^{-1} and at LNT, $\ln Z$=60–70, much higher than that in the conventional deformation. Consequently, the Zener-Hollomon parameter could be used for specifying plastic deformation process. One may reasonably anticipate that the higher the Z value (due to a higher strain rate and/or a low temperature), the more dislocations stored by deformation, and hence the smaller grains size achieved, as schematically illustrated in Fig.2(a) and 2(b).

With a substantial structure refinement below 100 nm, mechanical strength is expected to be elevated significantly. Previous experimental evidences have confirmed that the classical Hall-Petch relationship can be extrapolated to nano-sized grains (as small as 10 nm for Cu)[20]. Thereby, one may anticipate that yield strength or hardness of the DPD metals with larger Z parameters would be elevated relative to that at low Z. As shown in Fig.2(c), an increasing strength or hardness is expected with increasing Z parameter. This trend was proven in the bulk Cu sample processed by means of DPD of which the microstructure is refined into the nanometer scale consisting of nano-scale twins embedded in nano-grained matrix. Uniaxial tensile tests showed that the DPD Cu sample exhibits a tensile yield strength of about 600 MPa and an ultimate tensile strength of 633 MPa[14]. Both are much higher than those of SPD Cu samples (415, and 455 MPa, respectively[7]). In addition, the tensile elongation-to-failure of the DPD Cu is about 11%, which is comparable to that of the SPD Cu[14,7].

Plastic deformation of metals with high strain rates can be technically realized by using different loading routes such as dynamic loading, explosive loading, and electromagnetic pulse loading, *etc.* From a technical point of view, DPD processes are neither difficult nor complicated, and the processing costs will be low. A dynamic compression facility was recently developed in authors′ laboratory, which has been successfully employed in DPD processing of the bulk nanostructured Cu and Cu alloy specimens. Different DPD techniques are to be developed in the future for processing materials depending upon their dimensions and shapes. Bulk nanostructured materials or workpieces or parts with novel properties and performance for industrial applications depend upon development of the DPD techniques of industrial scale.

It should be noted that other grain refinement mechanisms are not excluded during DPD processes of metals and alloys. For example, some phase transformation kinetics is enhanced by plastic straining at high strain rates. As phase transformation facilitates grain refinement (such as in stainless steels[13]), high-strain-rate induced phase transformation provides an alternative grain refinement mechanism in the nanometer scale. Relative systematic investigations are needed to identify new grain refinement mechanisms.

Various physical and chemical properties, especially mechanical properties, are to be investigated systematically with the bulk nanostructured metal samples processed by DPD. Specimens in bulk forms are crucial for experimentally measuring the intrinsic properties (such as fracture toughness and fatigue properties), which in turn are important for understanding the intrinsic structure-property relationships in the nanometer scale. Extensive and intensive studies of these properties of nanostructured metals processed *via* using DPD are highly expected in the future.

Acknowledgements

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